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Calcic soils as indicators of profound Quaternary climate change in eastern Isfahan, Iran

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ABSTRACT

In the high plateau region of central Iran, there is both a lack of information on calcic soil processes and aspects of Quaternary paleoclimate. Thus, the research discussed here was undertaken as a first step to assess the utility of calcic soil research in the region. The properties and isotope composition of calcic soils was studied on a complex of alluvial fans, located about 50 km SE of the city of Isfahan. At least three geomorphic surfaces have formed during an interval from the middle Pleistocene to the Holocene. These geomorphic surfaces contain calcareous soils and paleosols. Interpretation of the pedogenic evidence indicates that there have been multiple periods of clay formation and carbonate accumulation over time, with an overall trend of increasing environmental aridity. Microscopy shows that overprinting is a major factor responsible for the accumulation of calcrete, suggesting the impacts of climate oscillations on the calcrete formation. Stable isotope composition of pedogenic carbonates in soils of differing ages is suggestive of a decrease in plant density and an increase in evaporation as soils become younger. This research highlights the utility of the morphology and isotope chemistry of calcic soils for constraining environmental change during the Quaternary.

1. Introduction

Soil is the product of biogeochemical alteration of parent materials, and is an important part of the Earth's critical zone (Amundson, 2005). Soils formed at different times in arid environments provide important information on rates of geochemical processes and, in some circumstances, provide data on changes in regional climate conditions over geological time (McDonald et al., 2003; Oerter et al., 2016).

The rates of calcic soil formation (e.g. Machette, 1985) and climate history (e.g. Dworkin et al., 2005; Zhou and Chafetz, 2009; Ebeling et al., 2016; Bayat et al., 2017a) have been topics of research. Because climate is one of the important factors of pedogenesis, soil-based paleoclimatic interpretations have grown in use during recent years (e.g. Pfeiffer et al., 2012; Oerter et al., 2016; Bayat et al., 2017a). Oerter et al. (2016), using the stable isotope composition of pedothems (pedogenic carbonate clast coatings), found changes in atmospheric circulation pattern over North America during marine isotope stage (MIS) 4. Based on pedologic features, Pfeiffer et al. (2012) detected alternating climate cycles in central Chile that extend back to at least MIS 11.

Calcic soils form via different pathways on calcareous (e.g. Rabenhorst et al., 1991; Bayat et al., 2017b) and noncalcareous (e.g. Monger and Daugherty, 1991; Whipkey et al., 2002; Achyuthan, 2003) parent materials. External sources of Ca ions (dust or Ca ions in rain water) are important for the development of pedogenic carbonates in noncalcareous parent materials. On other hand, pedogenic carbonates forms more quickly in calcareous parent materials due to in situ alteration of the relatively soluble calcareous minerals.

Calcareous soils and paleosols formed in geomorphic surfaces of differing ages provide information for discrete points or intervals in

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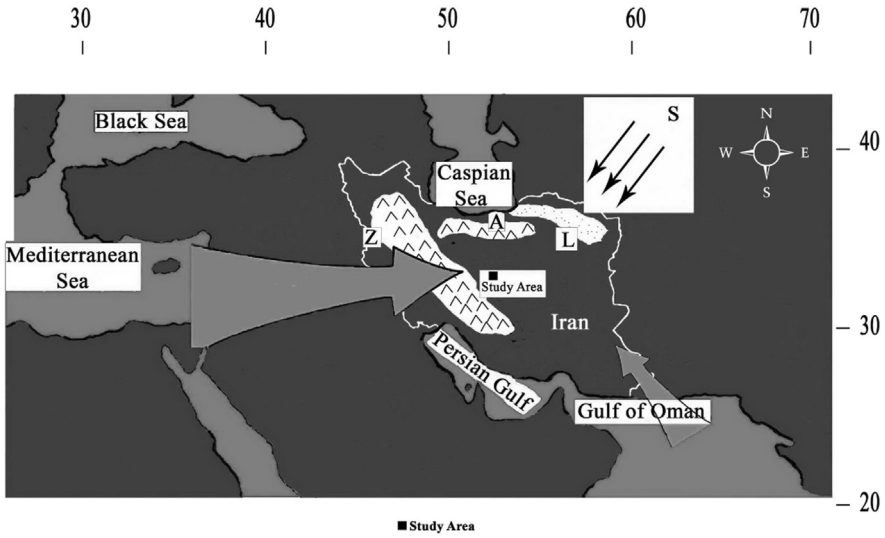

Fig. 1. Study area in central Iran; Z: Zagros Mountains, A: Alborz Mountains, L: loess deposits of northeastern Iran, S: Siberia High. Thickness of arrows shows the importance of moisture source from the Mediterranean Sea or the Gulf of Oman (monsoon) for Iran.

time (e.g. McDonald et al., 2003; Young et al., 2004; Bayat et al., 2017b). A geomorphic surface is defined as a mappable landscape element formed during a given time period. Surfaces are distinguished from one another by distinctive topographic features, weathering characteristics and pedogenic development (Christenson and Purcell, 1985; Bull, 1991; Schaetzl and Anderson, 2005). Geomorphic surfaces may be depositional, erosional or a combination of both (Schaetzl and Anderson, 2005). Geomorphic surfaces of alluvial fans are commonly used as the basis for chronostratigraphic studies of arid soils (e. g. McDonald et al., 2003; Bayat et al., 2017b) and the nature of pedogenic processes (Sauer et al., 2015).

Central Iran possesses a transitional climatic region, at the boundary between a westerly-dominated zone in north (from the Mediterranean and Black Seas) and a zone influenced by a sub-tropical High to the south. Thus, relative shifts in the strength of either of these systems should produce significant changes in the local climate. However, there is a considerable lack of information on aspects of Quaternary paleoclimate. Thus, the research discussed here was undertaken as a first step to assess the utility of calcic soils as a paleoclimate tool in the region.

2. Geographic setting

The field area chosen for this study is a complex of alluvial fans about 50 km SE of the city of Isfahan (Fig. 1). Geologically, the study area is situated in the Sanandaj-Sirjan tectonic zone, which is the most active tectonic zone of Iran (Aghanabati, 2004). The area is similar in structure to that of the Basin and Range Province of the USA (Karimzadeh, 2002; Bull, 1991). The alluvial fans drain a small mountain range along the southern margin of the Zayandehrud River valley. This river, like many in this region, is internally drained and enters the Gavkhoni playa 80 km SE of the field area (Figs. 1 and 2). Alluvial fans are major landforms in central Iran (e.g. Le Dortz et al., 2011; Jones et al., 2014), with great expanses of these features in the Isfahan region (Fig. 2).

The study area is affected by a rain shadow effect of the Zagros Mountains (Fig. 1). The Zagros Mountain range is separated from central Iran by the Zagros thrust fault. Due to interest in the rates of offset along this fault system, Le Dortz et al. (2011) have provided geochronological constraints on the ages of the alluvial fans in that region. While these sites are 250 km to the SE of the area studied here, the similar elevations and geological setting suggest that the work of Le

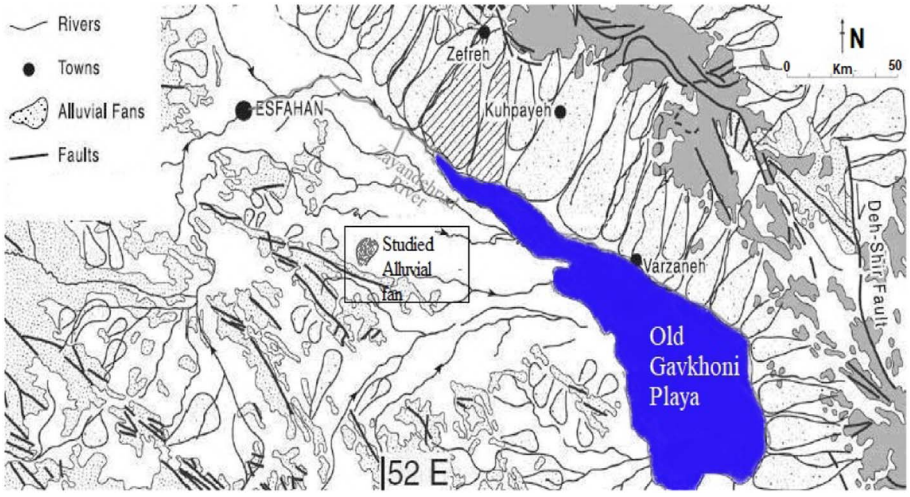

Fig. 2. Location of the alluvial fans in the eastern Isfahan region (after Jones et al., 2014). Notice the fault along the mountain front of the studied fans.

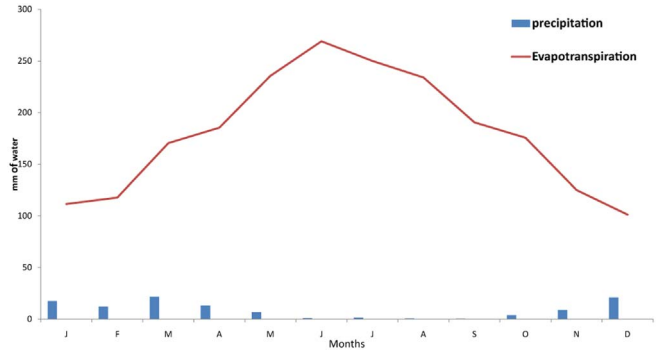

Fig. 3. Mean monthly precipitation and evapotranspiration in Isfahan (1968–2005).

Dortz et al. (2011) provide chronological constraints that may be useful to the region as a whole.

Cretaceous limestone and Jurassic shale are the dominant source material for the alluvial fans, and the fans thus consist of Quaternary sediments which are calcareous (Zahedi, 1976). Despite aeolian processes and sediments that exist in the eastern Isfahan, several lines of evidence suggest dust unlikely has a significant role as parent material for these calcic soils: (1) While aeolian processes and deposits (e.g. sand dunes and ripples) are common in the basin lowland and playas (Karimzadeh, 2002; Bayat, 2007), Av horizons representing significant accumulations of aeolian materials are not present in the soils studied here, or on other alluvial fans in eastern Isfahan (Karimzadeh, 2002). (2) Calcareous gravels within the alluvium provide a ready source of Ca for pedogenic carbonate and, (3) aeolian sediments in the eastern Isfahan region are mainly gypsiferous (Karimzadeh, 2002). Due to the limestone-rich alluvium, there is a large in situ carbonate reservoir to form pedogenic carbonate (McDonald et al., 2003). However, the role of addition of clay-sized dust in development of these soils cannot be excluded completely. Further studies must be carried out in the area to quantify dust fluxes and the role of dust in the development of the soils of the region.

3. Modern climate and vegetation of the eastern Isfahan, central Iran

The present climate of the study area is arid to hyper-arid, with a mean annual precipitation and temperature of about 120 mm and 15.2 °C, respectively. Mean annual potential evapotranspiration of the study area, calculated using the Blaney-Criddle method, is about 2166 mm (Fig. 3). The pedoclimate of the study area is characterized by aridic and thermic moisture and temperature regimes, respectively.

The climate of southwest Asia is affected by the Siberian High (in winter), the monsoonal Asian Low (in summer), eastward low-pressure systems from the Mediterranean (in non-summer seasons) (Fig. 1), and the Azors High (during dry seasons) (Cullen, 2005; Masoodian and Kaviani, 2008). The position and intensities of these systems over Iran changed during Quaternary (Kehl, 2009). Because central Iran is located in a transitional zone between northern westerlies and southeastern monsoons, this area may have been influenced by either enhanced westerlies and/or southeast monsoons during the Quaternary (Jones et al., 2014). The precipitation regime of southwest Asia is characterized by significant wintertime precipitation (Cullen, 2005). Westerlies shift storm tracks and cyclones from the Mediterranean into central Iran in the cold seasons (Masoodian and Kaviani, 2008). However, the eastward extension of the Azors High results in hot and dry conditions through the year in large parts of the southwest Asia, and especially in central Iran (Masoodian and Kaviani, 2008).

In order to evaluate present-day climatic conditions with respect to geomorphic and pedogenic processes, the soil-leaching index (SLI) and seasonality indices were calculated and used (Bull, 1991). The soil-leaching index is an estimate of annual water into soils, and is calculated using the annual sum of monthly precipitation minus monthly evapotranspiration for those months with positive values (Bull, 1991). Seasonality of Precipitation index (sp) is the ratio of average total precipitation of the three wettest consecutive months to the average total precipitation of three consecutive driest months (Bull, 1991). Seasonality of temperature index (st) is defined by Bull (1991) as the difference between the mean temperature of the hottest month and mean temperature of the coldest month.

The present-day climate is one where monthly potential evapotranspiration exceeds precipitation throughout the year (Fig. 3). Therefore, the SLI index for the study area is equal to zero. This suggest that there is little possibility for soil leaching to significant depths under present-day climatic conditions, and that many pedogenic features on pre-Holocene surfaces are possibly relict and the result of past leaching regimes and climatic conditions.

There is strong seasonality in temperature and precipitation in the study area, with seasonality values of precipitation and temperature being 60.6 and 28.8, respectively.

Ecologically, the current vegetation is composed of C_3 desert shrubs (shrubs which use the C_3 photosynthesis pathway) and C_4 grasses, with an overall dominance of C_3 plants (Khademi and Mermut, 1999). *Artemisia Sieberia* L., *Scariola orientalis* Boiss, *Launea spinosa* Forsk, *Euphorbia gedrosiaca*, *Anabasis aphylla* L., *Noea mucronata* and *Salsola nitraria* Pallas are the most important native plants of the study area (Khademi and Mermut, 1999).

Groundwater in the study area is deep, and there is no evidence for groundwater-created redoximorphic conditions.

4. Methodology

The sequence of alluvial fans and fluvial features were identified using aerial photographs, satellite images, and field surveys. Three surfaces were identified: Qf1, Qf2, and Qf3, oldest to youngest, respectively. Based on morphological comparisons to alluvial fans studied by Le Dortz et al. (2011) in central Iran, we tentatively correlate our Qf3 (Holocene to late Pleistocene alluvium) to their T1, our Qf2 to their T2 (107 ka (MIS 5c) to 412 ka (MIS 11)), and our Qf1 has no comparable identified landform in their field area, and probably is older than 412 ka. This suggests the geomorphic surfaces examined here span a time frame from the middle Pleistocene into the Holocene. Even without the work of Le Dortz et al. (2011), this tentative age assignment is reasonable based on comparison to research in other arid regions. We hypothesize that climate change may play a role in the processes of erosion and deposition that drive fan formation, as has been suggested for alluvial fan systems in SE Iran (Kober et al., 2013).

Dissection of geomorphic surfaces is a relatively rapid geomorphic response to tectonic changes and/or climatic change, and is a good indicator of relative ages within a given region (Ponti, 1985; Bull, 1991). The Dissection Index (DI), which is a quantitative geomorphic index to calculate the dissection of sediments by the character of contour lines of topographic maps, was calculated for the geomorphic surfaces based on the method of Ponti (1985). As geomorphic surfaces of the study area are very small, we were able to study only three contour lines on each fan. DI values were calculated using ILWIS 3.3 software.

Soil profiles representative of each surface were described using the methods of Soil Survey Staff (1999) and classified according to Soil Taxonomy (Soil Survey Staff, 2014a). In addition to bulk samples from each horizon, secondary carbonate samples were collected from representative pedons on geomorphic surfaces of the alluvial fans. For the soil on Qf1, samples were taken from the upper (80–86 cm), middle (102–112 cm) and lower (120–125 cm) depths of a Btk horizon which contain thin layers of Bt and Bk horizons. The morphology of pedogenic carbonates was described using to the method of Machette (1985). Soil pH was measured in a saturated paste and electrical conductivity (ECe)

of the soil samples was measured in the saturation extract.

The calcium carbonate equivalent (CCE) was determined by acid neutralization (Soil Survey Staff, 2014b). Organic carbon was determined by the Walkley-Black wet digestion method and multiplied by a standard conservation factor of 1.724 to convert organic carbon into organic matter content (Soil Survey Staff, 2014b). Particle size distribution was determined by the pipette method following the removal of organic carbon with 30% H_2O_2 and the addition of dispersants and shaking (Soil Survey Staff, 2014b). Due to the calcareous nature of the parent material, acid pretreatments were not made prior to particle size analyses. Soil color of air-dried samples was determined under natural light using the Munsell color chart.

X-ray diffraction (XRD) patterns for selected calcic horizons were obtained with a Bruker D8 Advance diffractometer using powdery bulk carbonates and Cu-ka radiation (35 kV, 25 mA).

Scanning electron microscopy (SEM) studies were undertaken using a Philips XL 30 SEM (voltage: 20 V, current beam: 65 μA). Samples were coated with gold before SEM analysis. Energy Dispersive X-ray Spectroscopy (EDX) was performed using a SERON AIS 2300 device. We use the terms micrite (crystal size < 4 μm), microsparite (5–20 μm) and sparite (> 20 μm) to describe size range of the observed carbonate crystals (Durand et al., 2010).

For isotopic analysis, ten microsamples (about 1 g) of pedogenic carbonates were collected from each calcic or petrocalcic horizon and the pedogenic carbonates were crushed, and mixed to produce the sample of the horizon. The samples were collected from below the depth of 25 cm to reduce the effects of CO_2 diffusion and evaporation on isotopic enrichment (Cerling, 1984; Cerling and Quade, 1993). The carbonate samples were reacted with 100% phosphoric acid to release CO_2 gas and the CO_2 was purified and analyzed using mass spectrometry at the University of California, Berkeley. Isotopic values were reported in the standard δ notation (Eq. 1) relative to Pee Dee Belemnite (PDB) standard (Hoefs, 2009) (Eq. 1).

$$\delta\,(‰) = [(R_{Sample}/R_{Standard}) - 1] \times 1000 \quad (1)$$

5. Results and discussion

5.1. Geomorphic surfaces of the alluvial landscape

Remote sensing interpretations (satellite images and aerial photographs) and field observations of the study area suggest two major generations of erosion and deposition. The first generation of fan deposits formed immediately along the mountain front (Qf1) (Fig. 4). Based on the existing remnants, these are steeply dipping surfaces (slope = 10.9%) that are highly dissected. The mean DI value for Qf1 is 1.52, which indicates severe dissection (Figs. 4 and 5). A major fault underlies the mountain front in the study area (Fig. 2), and this tectonic activity has had an influence on fan dissection in the Isfahan region (Jones et al., 2014). The next generation of fan deposits (Qf2, Qf3) covers most of the surface of the piedmont (Fig. 4A). The Qf2 surface has a lower gradient than Qf1 (Table 1). The mean DI value of this surface is about 1.18, and it has a slope of 3.44%, which is much gentler than Qf1 (Table 1). Qf3 is a geomorphic surface impacted by modern deposition/erosion during flash-flood events, and has a mean of DI value of 1.24. This alluvial fan system extends downslope to the nearby microplaya (Fig. 4A) which is enriched by gypsum (Bayat, 2007). Near the terminal end of the fan system, there are relict surfaces or terrace remnants (Fig. 4A). A prior stratigraphic study of these surfaces showed they are relicts of an old lacustrine landform with a gypsiferous soil at the surface (Bayat, 2007). These remnants have subsequently eroded and have been partially buried by Qf3 fan sediments, and now have limited outcrops.

The amount of dissection of a geomorphic surface reflects relative ages (Ponti, 1985). Work on calcic soils in the Mojave Desert have shown that soil hydraulic conductivity tends to decline with increasing

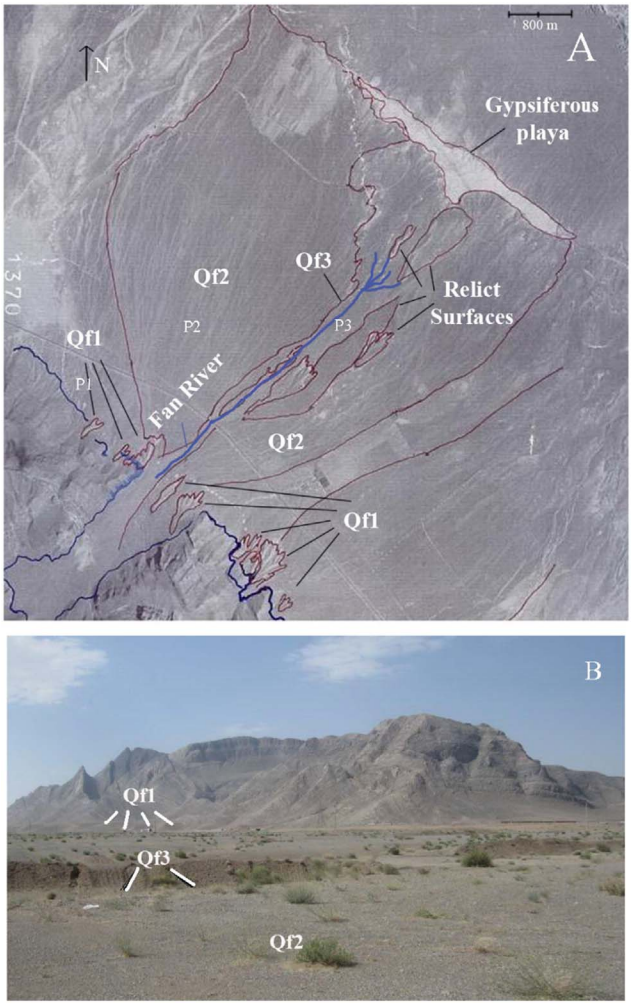

Fig. 4. (A) Aerial photograph of the geomorphic surfaces of the alluvial fan system. (B) Ground photo of the geomorphic surfaces of the fan in eastern Isfahan. Note that Qf1 is located near the mountain front, and that very scarce desert shrubs (C_3 plants) are the only vegetative cover. Qf3 has formed along the active fan channel, and is slightly below eye level. The locations of representative pedons are characterized by P1, P2 and P3.

soil age (due to the formation of dust-rich V horizons and other pedogenic changes), which in turn facilitates erosion and the development of drainage networks (Young et al., 2004). DI values for the Qf2 and Qf3 geomorphic surfaces are similar, with a slightly larger value for Qf3 surface than Qf2 (Fig. 6). The somewhat higher values for the Qf3 surface likely reflects the surface roughness associated with its active deposition systems, features that tend to level out once active fans are abandoned and begin undergoing soil development. This suggests that DI values are more appropriate for dating or mapping purposes on inactive surfaces.

We have no direct geochronological controls on the fans except for their relative ages. Recently, Le Dortz et al. (2011) used a combination of cosmogenic radionuclides (CRN) and optically stimulated luminescence (OSL) to date alluvial fans in essentially the same geomorphic province about 250 km to the SE. Due to high resolution digital imagery (Digital Globe) available at both locations on Google Earth, we feel confident about tentatively correlating the fans/ages at their site to the ones examined here (Table 1). The data suggest that the chronosequence examined here spans from essentially modern to ~412 Ka for deposits of Qf3 and Qf2, respectively. A surface equivalent to Qf1, whose remnants are found only as isolated exposures near the mountain fronts (Fig. 4A), was not identified in their study, which was located considerably down the drainage system and far away from

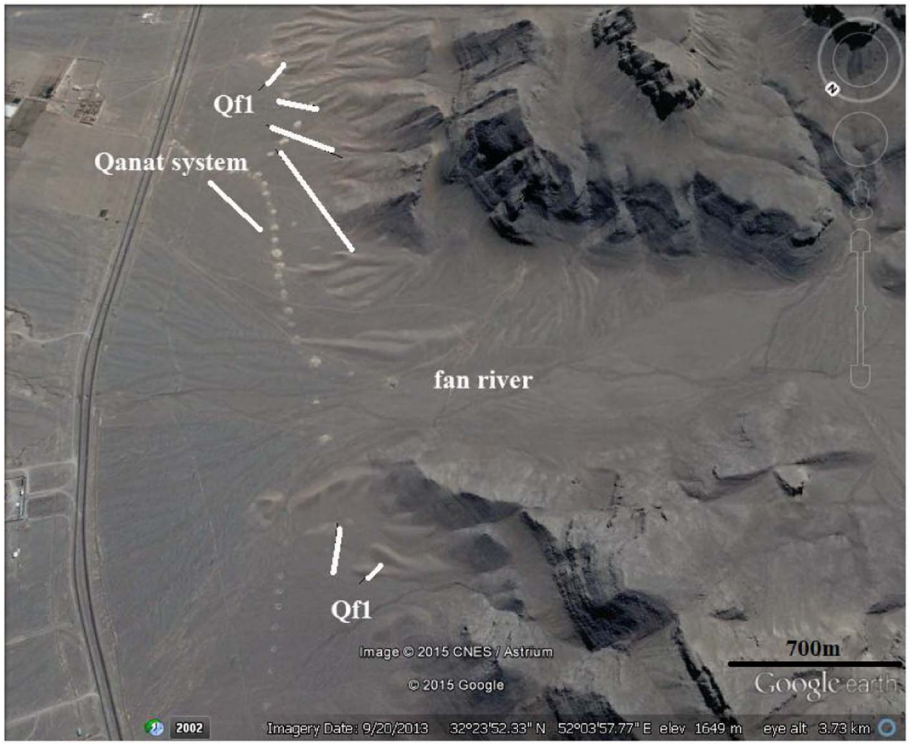

Fig. 5. Old dissected alluvial fans (Qf1) on the mountain front. Note that the active fan channel transitions to a braided stream further down the fan. The Qanat system was created to extract water from the fan and transport it to lowlands.

Table 1
Geographical settings of the studied geomorphic surfaces.

Geomorphic Surface	Longitude	Latitude	Altitude (m)	Slope (%)	Parent material	Age (ka)
Qf1	52° 03 E	32° 23 N	1627	10.9	Calcareous alluvium	> 412 ka
Qf2	52° 04 E	32° 25 N	1587	3.3	Calcareous alluvium	107–412
Qf3	52° 05 E	32° 24 N	1553	1.8	Calcareous alluvium	0–107

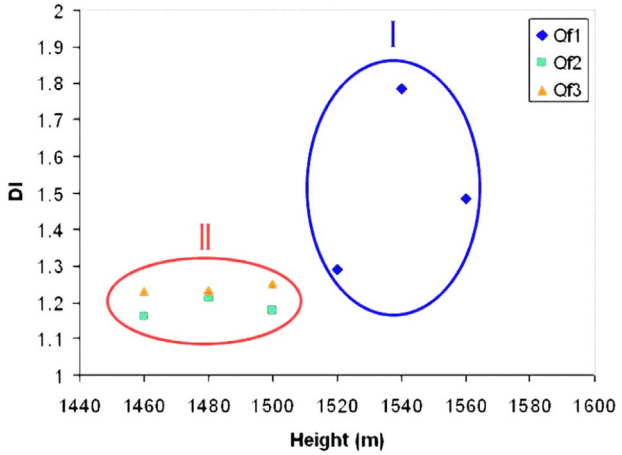

Fig. 6. Dissection Index (DI) values for the studied geomorphic surfaces. The differing colors group generations of fan deposits (I vs. II).

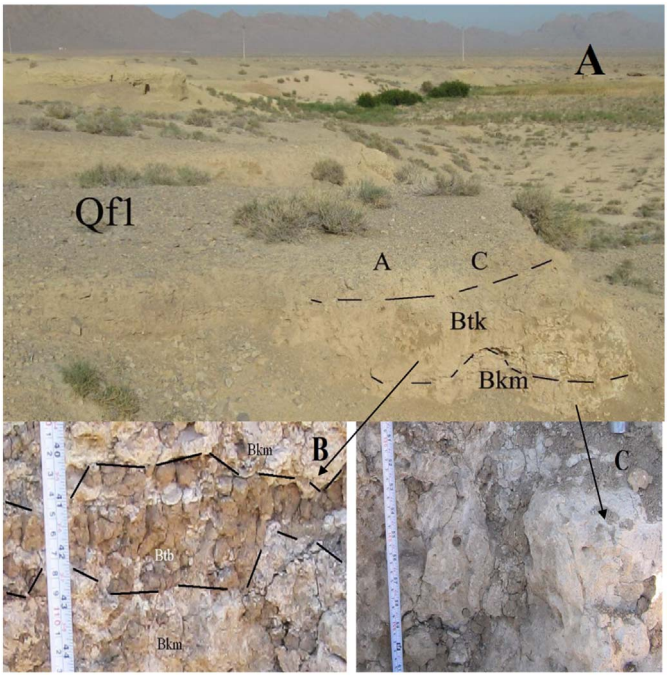

Fig. 7. (A) Pedogenic development of a profile representative of pedon Qf1. (B) Close up of Bk and Bt horizons in the middle part of Pedon 1. (C) Massive calcrete in the bottom of Pedon 1.

mountainous sources. In eastern Isfahan, fan abandonment has occurred from mid Tertiary to the early Holocene (Jones et al., 2014). Fan aggradation and abandonment processes are generally attributed to changes in global climate in the Mojave Desert of California (McDonald et al., 2003) and SE Iran (Kober et al., 2013). This it is likely that the second generation of geomorphic surfaces (Qf2-Qf3) in eastern Isfahan have formed in a stable to active tectonically settings, but also under the influence of climatic changes. Regardless of the relative importance of tectonics vs. climate, the different aged surfaces provide soils that capture different time intervals of soil formation.

5.2. Soils properties and formation

The paleosol of Qf1 geomorphic surface (Pedon 1) is a truncated soil

Table 2
Morphological, physicochemical and isotopic properties of studied pedons.

Horizon	Depth (cm)	Dry color	pH	ECe (dS/m)	OM (%)	CCE (%)	CMS	$\delta^{18}O$ (PDB, ‰)	$\delta^{13}C$ (PDB, ‰)	Sand (%)	Silt (%)	Clay (%)	Texture	C/SS
Qf1: Argic Petrocalcids, 32°24′53″N, 52°02′26″E														
A	0–25	10YR6/4	8.0	4.7	0.49	41.4	–	–	–	54.1	21.2	24.7	SCL	0.32
C	25–42	10YR6/3	8.3	12.6	0.09	29.5	–	–	–	60.3	15.9	23.8	SCL	0.31
2Bkmb1	42–80	7.5YR6/4	7.4	8.8	0.18	71.8	III	− 7.78	− 2.77	18.7	59.7	21.6	SiL	0.27
2Btb2	80–83	7.5YR5/4	7.5	5.9	0.26	43.5	–	–	–	26.8	19.4	53.8	C	1.16
2Bkmb2	83–86	10YR6/4	7.5	3.3	0.07	92.6	III	− 9.01	− 3.21	16.3	58.6	25.1	SiL	0.33
2Bkmb3	102–107	10YR5/4	7.6	3.4	0.06	81.6	III	− 7.57	− 1.95	12.9	63.3	23.8	SiL	0.31
2Btb2	107–112	7.5YR5/4	7.5	8.9	0.45	28.6	–	–	–	8.7	35.0	56.3	C	1.28
2Btb3	120–125	7.5YR6/3	7.5	9.5	0.29	35.9	–	–	–	9.7	56.1	34.2	SiCL	0.52
3Bkmb	+ 125	10YR6/4	7.5	8.7	0.07	82.6	IV	− 8.40 − 8.02	− 4.15 − 3.04	27.4	58.7	13.9	SiL	0.16
Qf2: Typic Calciargids, 32°24′45″N, 52°04′32″E														
A	0–17	10YR6/3	7.6	5.2	0.08	31.2	–	–	–	73	14.6	12.4	SL	0.14
Btk	17–48	7.5YR4/6	7.3	9.4	0.15	27.2	I	− 1.12 − 1.94	+ 0.58 − 0.24	48.6	20.5	30.9	SCL	0.44
2C	48–73	7.5YR5/4	7.4	10.2	0.08	29.6	–	–	–	63.2	11.9	24.8	SL	0.33
3Btk	73–125	5YR4/4	7.4	5.4	0.18	31.6	I-II	− 3.05	+ 1.43	34.4	24.4	41.1	C	0.69
4C	+ 125	7.5YR5/4	7.3	4.8	0.07	25.4	–	–	–	56.3	15.2	28.5	SCL	0.39
Qf3: Typic Torriorthents, 32°25′02″N, 52°05′55″E														
A	0–6	10YR6/4	7.9	1.5	0.51	42.8	–	–	–	69.1	16.6	14.2	SL	0.12
C1	6–45	10YR5/3	8.1	1.7	0.45	32.8	–	–	–	68.7	13.8	17.5	SL	0.21
C2	+ 45	10YR5/4	7.9	1.6	0.16	29.8	–	–	–	70.8	12.2	17.0	SL	0.20

OM: Organic material, CCE: calcium carbonate equivalent, CMS: Carbonate morphology stage, C/SS: Clay/Sand + Silt, SCL: Sandy clay loam, SiCL: Silty clay loam, SiL: Silt loam, SL: Sandy loam.

which is covered by younger deposits (Fig. 7). This pedon consists of two main parts: (1) an immature surficial soil (A and C horizons) and (2) the underlying truncated paleosol (Fig. 7). The buried soil retains a sequence of calcic, calcic-argillic and petrocalcic (calcrete) horizons (Fig. 7). The upper, younger section has formed within calcareous gravelly materials without significant pedogenic development. The horizons have high pH values which indicate an accumulation of bases reflective of the present day low leaching rates (Table 2). There is more organic carbon and sand in the overlying soil than within the buried soil. The surficial part of the soil reflects a past instability of the landscape and recent (ephemeral) floods on the fan surface. The age of the overlying unit must be relatively recent due to the minimum weathering that has occurred. This overlying material (A and C horizons) is characterized by angular calcareous clasts, with no observable secondary weathering. In contrast, the lower part of Pedon 1 contains strongly contrasting pedogenic features. These horizons indicate multiple stages of carbonate and clay accumulation, particularly through the observation that some clay rich structural units are low in, or devoid, of carbonate, which nonetheless has largely invaded the previous non-calcareous layer (Fig. 7). These pedogenic processes took place over a considerable period of time on a stable landscape that was probably influenced by much more water and associated denser vegetation. The horizons of clay accumulation are thin (due to partial truncation), and contain low carbonate and high clay content. These horizons also contain more organic matter (Table 2) probably in response to stabilization of organic matter by clay particles. These argillic horizons have clay coatings on peds, and contain large amounts of pedogenic smectite in the clay fraction (Bayat et al., 2017b). Because argillic horizon formation require decarbonation and large amounts of leaching and time (Sauer et al., 2015; Ebeling et al., 2016), these horizons are strong evidence for a past subhumid paleoclimate in the study area. The leaching depth of soil carbonate is somewhat sensitive to soil age, though rainfall is the most important factor in carbonate leaching depth in soils (Egli and Fitze, 2001). The calcrete horizons of this pedon contain carbonate content between 71.8 and 92.65%, which is similar to the global average of carbonate content of calcretes (79.2%) (Goudie, 1972). The field morphologies of these horizons consist of coalesced nodules and massive calcrete with thin laminae (stages III and IV according to Machette, 1985). The morphologies and total quantity of pedogenic carbonates are considered indicators of relative soil age (Machette, 1985; Zamanian et al., 2016).

The Qf1 paleosol has significant quantities of pedogenic smectite (Bayat et al., 2017b), reflecting considerable past chemical weathering. Decarbonation is the most important prerequisite for the lessivage process to occur in calcareous soils (Schaetzl and Anderson, 2005), and rainfall is the most important factor for carbonate leaching (Egli and Fitze, 2001). In general, the accumulation of carbonates and the chemical weathering of silicates, and accumulation of clays, are a set of incompatible processes, and suggest that the soil has experienced declining moisture during its exposure (e.g. Ebeling et al., 2016).

Different mechanisms have been proposed for mineral dissolution and secondary mineral formation in calcic soils (e.g. pressure solution and crystallization force (Robins et al., 2012)). Pressure solution is the dissolution of grains due to lithostatic pressure, and is restricted to grain-to-grain contacts (Robins et al., 2012). The force of crystallization is caused by the growth of secondary minerals, and a subsequent increase in the solubility of interacting grains (Monger and Daugherty, 1991; Robins et al., 2012). In calcic horizons, quartz grains may be affected by the force of calcite crystallization, increasing the release of ions (especially Al, Si and Mg) into the soil solution. This has mechanism has been proposed to be involved in the formation of palygorskite and sepiolite in calcrete horizons (Robins et al., 2012), as well as fibrous clay minerals (Monger and Daugherty, 1991; Achyuthan, 2003). The petrocalcic horizons in Pedon 1 contain pedogenic palygorskite (Bayat et al., 2017b). Whatever the mechanism that aided the formation of these minerals, the high soil pH and abundant Ca, Mg, and Si are ideal conditions for the formation of this mineral.

Pedon 2, which formed on the Qf2 surface, is a relict soil consisting of a sequence of fluvial materials that have been altered to argillic-calcic horizons (Btk horizons). The carbonates occur as discontinuous and continuous coatings on pebbles and peds (stages I and II) in Btk and 2Btk horizons, respectively. These carbonates (on the surface of calcareous clasts) support the hypothesis that in situ weathering of calcareous pebbles is a major source of Ca for genesis of pedogenic carbonates. The sediment accumulated during a past landscape instability, where rate of sediment supply by floods (partially from the Qf1

surfaces) exceeded the rates of soil formation. Landscape stability later ensued, and soils have formed on stable surfaces. Textures of Pedon 2 are all loamy, except for the 3Btk horizon which has a clay texture (Table 2). This horizon is also characterized by a reddish hue (5YR) due to rubification, reflecting pedochemical weathering and the formation of hematite (Pfeiffer et al., 2012). The red argillic horizon of this pedon (3Btk-Qf2) is characterized by clay coatings covered by calcite crystals (Bayat et al., 2017b). The production of Btk horizons in the soils of northwest Spain and central Chile has been attributed to changes in climate from humid, to subsequent arid, conditions (Alonso et al., 2004; Pfeiffer et al., 2012).

In summary, the physicochemical properties of Pedon 2 indicate the effects of a calcareous parent material and limited moisture availability. The soil has less total clay than Pedon 1, but does contain clay enriched Btk horizons. Thus, these horizons contain signals from past weathering processes under higher leaching conditions. Thus, these soils, like Qf1, can be considered polygenetic paleosols (Schaetzl and Anderson, 2005). The last phase of carbonate accumulation has occurred as powdery accumulations on ped surfaces and clasts (stage I).

Pedon 3 is the soil examined on the youngest Qf3 geomorphic surface. This soil consists of an A horizon that overlies C horizons. The weak pedogenic development is due to its young age, landscape instability, and the current arid climate. In comparison to the other soils, this profile has a lower salinity, probably due to its coarse texture and semi-frequent flooding by active streams of the fan. There is also more organic carbon is in this soil (Table 2), the result of more moisture availability and plant productivity. Carbonate content in this profile decreases with depth (Table 2) due to addition of fresh calcareous materials to the soil surface by active streams. A high percentage of sand in the whole profile, in comparison with the other soils, is another indicator of its young age and weak weathering conditions.

The clay/silt ratio has been suggested as a simple index for silicate weathering and clay formation (Sauer et al., 2015). In the chronosequence examined here, we found a relationship between relative ages of soils and clay/sand + silt (C/SS) ratio rather than clay/silt ratio, probably due to the fluvial nature of the parent material and the low silt content (Table 2). As might be expected, the highest value of C/SS ratio is observed in the Bt horizons of Qf1 (Fig. 8). Clay mineralogical analyses confirm the formation of pedogenic smectite in this Bt horizon (Bayat et al., 2017b). The lowest value for the C/SS index is in the A horizon of Pedon 3 (Fig. 8), which is similar to the values for the C horizons of Qf1 and Qf2.

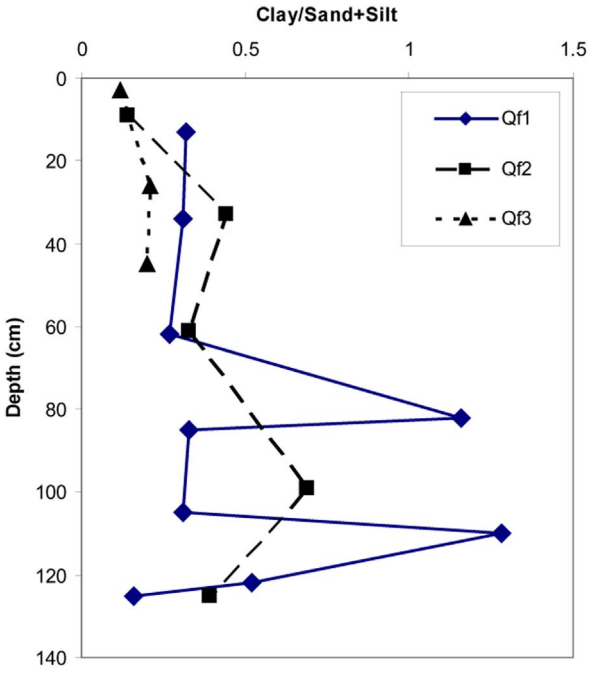

Fig. 8. Clay/sand + silt ratio for the studied soils.

The 3 soils, from oldest to youngest, classify as Argic Petrocalcids, Typic Calciargids, and Typic Torriorthents based on Soil Taxonomy (Soil Survey Staff, 2014a) and emphasize and reflect the decline in carbonate and clay accumulation with age (Fig. 9). However, in the soils of Qf2 and Qf3, the carbonate content decreases with depth (Fig. 9), due to additions of fresh calcareous materials to the surface of the fan.

In summary, the soils show that there was once enough environmental moisture to remove calcareous material from the horizons and form silicate clay in the two oldest soils, though the degree of weathering is greatest in the oldest soil. The morphologies and amounts of secondary carbonates are in agreement with their relative age (Table 2). The overlay of carbonate over secondary clays points to climatic oscillations, and the inference that the present climate is a carbonate forming environment, and that condition must have been different in order to form Bt horizons in the past.

5.3. Mineralogy and submicroscopy of carbonate accumulation horizons

The crystalline forms of pedogenic carbonates reflect interactions between the lithosphere, the biosphere and the atmosphere (Durand et al., 2010; Zamanian et al., 2016).

Diffractograms of carbonate rich horizons in Pedon 1 (Fig. 10) reveal a dominance of calcite (diagnostic peaks: 3.82, 3.02, 2.49, 2.27, 2.09, 1.91, 1.87 Å), especially the high intensity peak of 3.02 Å (Graf, 1961; Kraimer et al., 2005). There is no evidence for dolomite or gypsum, probably due to higher solubility of sulfate and relatively low Mg concentrations (Goudie, 1972).

Diffractograms of calcic horizons in Pedon 1 reveal quartz (peaks at 3.33 and 1.59 Å), which is probably derived from the host parent material. Quartz is the second most abundant constituent in most calcretes (Goudie, 1972). In general, the diffractograms are very similar to those of pedogenic carbonates that developed in limestones in southern New Mexico (Kraimer et al., 2005).

Geochemical properties of the carbonates here indicate low ratios of Mg/Ca, and a dominance of Ca and Si ions (Bayat et al., 2017b), which is agreement with X-ray mineralogy. The lack of a 3.00 Å peak associated with high Mg calcite indicates its absence in these soils (Whipkey et al., 2002; Zhou and Chafetz, 2009). The characteristic peak d(104) (i.e. 3.02 Å) of carbonates (Fig. 10) is narrow and symmetrical, which is also representative of low-Mg calcite (Zhou and Chafetz, 2009). In summary, mineralogical data and EDX spectra reflect low-Mg calcite in Pedon 1.

Photomicrograph 11 shows micrite calcite crystals in nodules in the uppermost calcic horizon of Pedon 1 (2Bkmb1). There are micritic rhombohedral calcite crystals (Fig. 11A) in this horizon. Micritic to microsparitic calcite with rhombohedral habit, have been interpreted as the initial stages of carbonate development (Zhou and Chafetz, 2009). In general, micrite crystals of calcite indicate rapid precipitation (Alonso et al., 2004), minimal recrystallization (Cerling, 1984) and a pedogenic origin (Zhou and Chafetz, 2009). Calcic horizons containing micrite crystals are considered suitable for isotopic studies (Cerling, 1984) and paleoreconstructions (Dworkin et al., 2005).

Photomicrographs of the underlying calcrete in Pedon 1 (3Bkmb) reveal two separate calcite textures (Fig. 11B, C): (1) sparitic rhombohedral crystals with sharp edges and crystal intergrowths and (2) calcite cement which filled rhombohedral crystals of calcite. The sparitic rhombohedral crystals show sharp edges, crystal intergrowth phenomena, and a lack of abrasion from mechanical processes (Fig. 11C). These features suggest authigenic (pedogenic) formation of these crystals.

The texture of the matrix and its porosity are important factors in the growth of pedogenic calcite crystals (Wieder and Yaalon, 1974; Zhou and Chafetz, 2009). Porous groundmass assists in the formation of

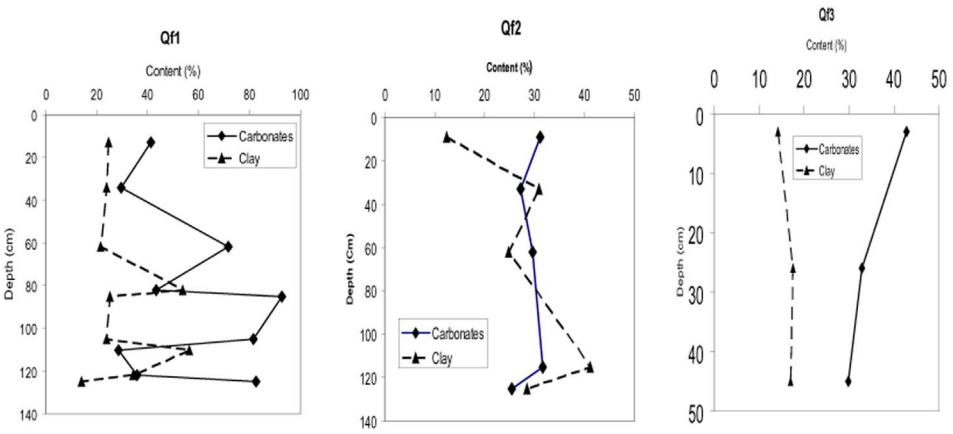

Fig. 9. Carbonate and clay content of the soils with depth.

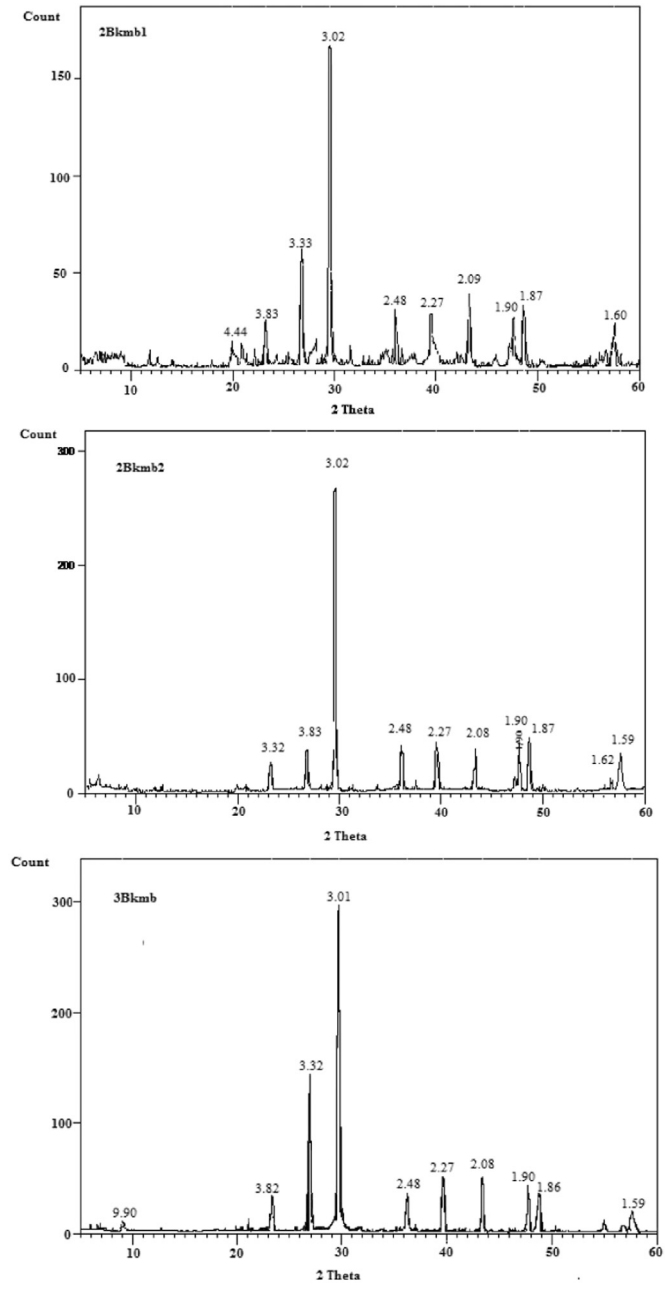

Fig. 10. Diffractograms of carbonate accumulation horizons, d-spacing numbers are in Å.

calcite crystals with rhombohedral habits (Zhou and Chafetz, 2009). Rhombohedral crystals are present in both the calcic horizon (R in Fig. 11A) and in the underlying calcrete (Fig. 11B, C) of Pedon 1. Rhombohedral crystals in the calcrete, however, are larger with sharper boundaries. Microsparitic to sparry calcite crystals indicate advanced stages of calcite growth and accumulation (Zhou and Chafetz, 2009) (Table 2).

Rhombohedral crystals in the calcrete are covered by, or embedded within, micritic calcite crystals (Fig. 11B, C). SEM observations suggest multiple phases or episodes of calcrete development: (1) the formation of rhombohedral crystals under stable and long-term conditions and a (2) penetration of micritic calcite crystals into rhombohedral crystals. The process of penetration and precipitation of younger carbonates into older ones is called overprinting (Deutz et al., 2002; Monger et al., 2009), due to several climatic episodes during soil development (Cerling, 1984; Monger et al., 2009). The calcrete in Qf1, stage IV with a high carbonate content (about 83%), is a likely candidate for overprinting. Somewhat similar calcretes studied by Deutz et al. (2002) in New Mexico also revealed overprinting. The formation of calcite during different environment periods may have an effect on the isotope composition of the overall carbonate, which is discussed below.

Calcite crystals in 3Btk horizon of Qf2 consist of micrite and microsparite calcite crystals, and lack rhombohedral crystals (Fig. 11D, E) and are partly covered by palygorskite fibers (Bayat et al., 2017b).

5.4. Stable isotope geochemistry

5.4.1. Stable oxygen and carbon isotope geochemistry

The oxygen isotope composition of pedogenic carbonate is largely determined by that of the soil water, which is derived from precipitation and possibly modified by evaporation enrichment of the remaining soil water in ^{18}O (Cerling and Quade, 1993; Amundson, 2005; Sheldon and Tabor, 2009).

The $\delta^{18}O$ values of precipitation depend on the storm source, the amount of precipitation, and the air temperature (Rozanski et al., 1992). The nearest station which monitors the $\delta^{18}O$ value of precipitation is in Tehran, which is north of the study area but has a similar elevation and comparable rainfall patterns. The $\delta^{18}O$ value of precipitation in Tehran varies from nearly 0 in the summer month of August, to as a low as − 9‰ in December (Fig. 12). Since in Tehran, as in Isfahan, most of the precipitation occurs in the winter from Mediterranean origins, both Tehran and Isfahan are shown on global compilation maps to have average precipitation values of roughly − 4 to − 6‰ SMOW (Mook, 2001). Assuming a mineral formation temperature of 15 °C, this implies pedogenic carbonates (on the PDB scale) should be between − 4 and − 6‰.

The $\delta^{18}O$ values of pedogenic carbonates in the Qf1 soil ranges from − 7.4 to − 9.01‰, much more negative than values expected from the

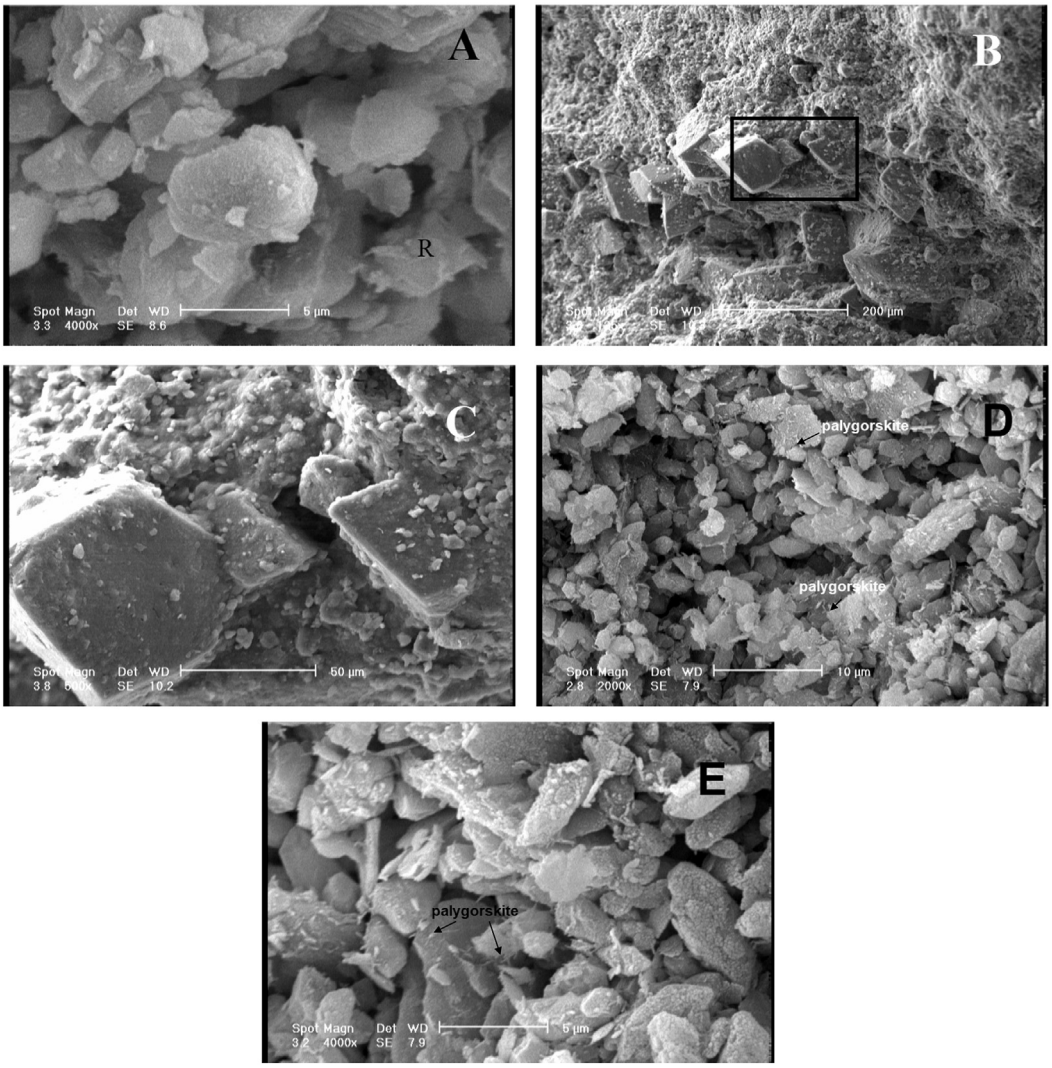

Fig. 11. Submicroscopy of the studied carbonates. (A) micrite and microsparite calcic crystals in calcic horizon in pedon 1 (Bkmb-Qf1). Note to rhombohedral calcite crystal (R). (B) Rhombohedral calcite crystals covered by micritic calcite crystals in massive calcrete of pedon 1. (C) Enlarged picture of rhombohedral crystals with sharp edges, crystal intergrowths phenomenon and lack of abrasion. (D) and (E) micrite and microsparite calcite crystals in calcic crystals of Btk horizon of pedon 2. Note the palygorskite fibers on the surface of calcite crystals of pedon 2.

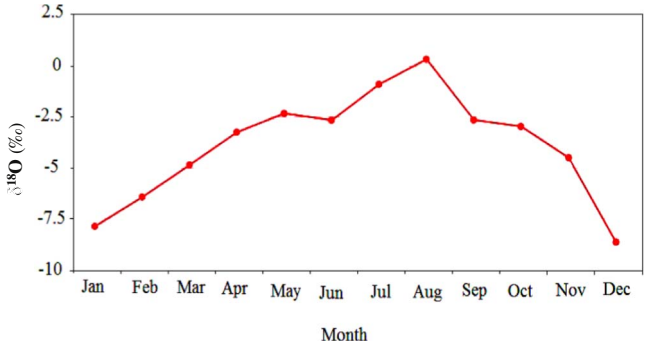

Fig. 12. Monthly variations in $\delta^{18}O$ values in meteoric water of Tehran. (after IAEA, www.iaea.org).

present rainfall values. In contrast, the Qf2 soil carbonate has $\delta^{18}O$ values that approach the predicted modern values, but that are slightly more positive (Fig. 13A). While we do not have isotope data for the more surficial horizons, there is a trend of increasing $\delta^{18}O$ values with decreasing depth in soil Qf2 (Fig. 13A), consistent with the effects of evaporative loss of water from the soil surface and subsequent isotopic enrichment (Achyuthan et al., 2007; Ebeling et al., 2016). Achyuthan et al. (2007) found that $\delta^{18}O$ values of soil moisture underwent significant O isotope enrichment by evaporation in the Thar Desert of India. Evaporation can result in enrichment of $\delta^{18}O$ value of soil carbonates + 5to + 7‰ (Sheldon and Tabor, 2009).

Paleotemperature reconstruction based on models presented by Dworkin et al. (2005), indicates that temperature was between 7.0 and 10.5°C colder during calcite formation in Qf1 geomorphic surface (Bayat et al., 2015). However, because several factors (e.g. change in rainfall source and amount, seasonality, evaporation and paleotemperatures) can all affect the O isotopic composition of pedogenic carbonate (Cerling and Quade, 1993; Amundson, 2005; Dworkin et al., 2005), caution must be taken in the estimation of paleotemperatures based on O isotopes. The $\delta^{18}O$ values of pedogenic carbonates of Pedon 2 are not suitable for paleotemperature reconstruction because they appear highly affected by evaporation.

The $\delta^{13}C$ values of soil carbonates are directly controlled by the isotopic composition of soil CO_2 respired by plant roots and organic matter decomposition (Cerling, 1984; Cerling and Quade, 1993). The $\delta^{13}C$ values of the carbonate in both soils is relatively enriched in ^{13}C, but the Qf2 soil is up to 5 ‰ more enriched than the Qf1. Presently, the local flora is largely dominated by C_3 plants. Based on isotopic

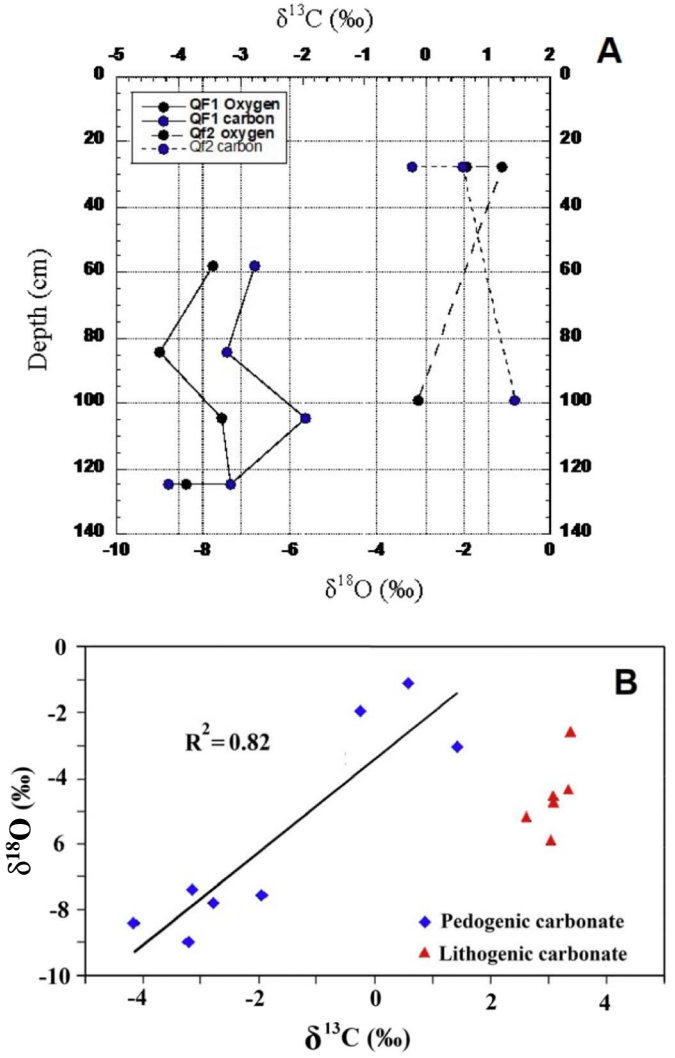

Fig. 13. Carbon and oxygen isotope values of studied carbonates. (A) Variation with depth for pedons 1 and 2. (B) Positive correlation for the studied pedogenic carbonates and isotopic values of parent materials.

composition of organic carbon in the soils of the study area, Khademi and Mermut, 1999 estimated that about 63% of the native plants in eastern Isfahan use the C_3 photosynthesis pathway. There is presently a scarce desert shrub cover (Fig. 4B and 7A) that provides low landscape stability (Dixon, 2013). Low plant density is correlated with low soil respiration rates, and an enrichment of ^{13}C in soil CO_2. It thus seems that shifts in paleovegetation density may be the key driving mechanism for the shifts in soil carbonate $\delta^{13}C$ values over time. The present warm and arid conditions might be expected to be an extreme in terms of aridity. The simplest interpretation of the C isotope data in the soils is that the carbonates reflect the relative proportion of biological CO_2 produced by respiration or decomposition of C_3 plants ($\sim -25‰$) to atmospheric CO_2 ($-6‰$ in preindustrial times) in the soil atmosphere. C_4 plants variations can have significant impacts on C isotopes in sub-humid to humid regions (e.g. Jiamao et al., 1997). However, C_4 plants are dominated by warm season grasses, requiring summer precipitation. While both soils reflect a significant fraction of atmospheric CO_2, there is a sharp increase in C isotope values from Qf1 to Qf2. A decline in biological CO_2 is consistent with a decline in plant cover. Thus, the two soils suggest a general decline in plant abundance over what might be a 500 Ka span of time. The decline in plant cover is consistent with the $\delta^{18}O$ values of the carbonate: declining plant cover leads to greater evapotranspirational losses of soil water and great enrichment of ^{18}O (Ebeling et al., 2016).

We note that some lithogenic carbonates may have been incorporated into the calcite of the youngest soil Qf2, though an effort was made to collect large pedogenic material. However, the measured C and O isotope values of lithogenic carbonate (Fig. 13B) is distinctively different than soil calcite samples.

What is especially interesting is the very negative $\delta^{18}O$ values of carbonate in the Qf1 soil, suggesting average precipitation values when the carbonate formed, as low as -8 to $-9‰$. A very simple interpretation is that the region received significantly greater influxes of winter rainfall when the Qf1 soil was forming, which additionally facilitated greater plant cover (and more negative $\delta^{13}C$ values). This interpretation is in excellent agreement with developed argillic horizons in Pedon 1 and occurrence of rhombohedral calcite crystals (Fig. 11B, C), which form in solutions with lower concentration of solutes (Zamanian et al., 2016). We are unaware of how circulation conditions in the mid-Pleistocene might have differed from today which could have delivered rainfall from different sources (e.g. Oerter et al., 2016), but at a minimum the Qf1 soil indicates the region had greater plant density, presumably more rainwater to support the flora, and more precipitation derived from cooler months than presently occurs. Moist conditions in northern and central Iran during pluvial periods of Quaternary are mostly ascribed to decreases in temperature and evaporation rates (Bobek, 1963; Krinsley, 1970) and southward shifts of westerly depressions (Kehl, 2009; Bayat et al., 2017a).

5.4.2. *Correlation between $\delta^{13}C$ and $\delta^{18}O$*

There is a correlation between the O and C isotopic values of the pedogenic carbonate, while lithogenic carbonates (parent materials) do not show any trend (Fig. 13B). A positive correlation between $\delta^{13}C$ and $\delta^{18}O$ values was also observed for carbonates from central Iran (Khademi and Mermut, 1999), northeastern Iran (Bayat et al., 2017a), and the Thar Desert of India (Achyuthan et al., 2007).

The positive correlation of C and O isotopes tends to occur since low vegetative cover (and respiration) tends to facilitate soil water evaporation (Cerling and Quade, 1993; Achyuthan et al., 2007), resulting in a positive correlation of $\delta^{13}C$ and $\delta^{18}O$ values. Overall, the sequence of soils and the carbonate isotope values are suggestive of declining plant densities, and precipitation O isotope values in the past that are much more negative than those of today, a trend due to circulation changes and/or increased evaporation rates.

6. Conclusion

This initial study of calcic soils on alluvial fans in Central Iran is suggestive of a large climate change over Quaternary times and the potential for pedogenic carbonates to provide greater information about the region's climate and geological history. On alluvial fans ranging from essentially Holocene to the mid-Pleistocene, we find pedogenic evidence for a shift from clay-forming environmental conditions ($\sim >$ 500 mm rainfall per year) to those that favor carbonate accumulation ($<$ 500 mm per year) (e.g. Egli and Fitze, 2001; Retallack, 2005). In addition, the stable isotope composition of pedogenic carbonate, both O and C isotopes, becomes considerably enriched in heavy isotopes as soils become younger. Carbon isotopes can be most simply interpreted as representing a steep decline in plant cover, likely caused by greater aridity. The O isotopes of the younger ($<$ 412 Ka) carbonates reflects evaporative enrichment of soil water in ^{18}O. However, the older carbonates (possibly $>$ 412 Ka) reflect significantly ^{18}O-depleted precipitation relative to that which occurs today.

This reconnaissance work supports the utility of more extensive stable isotope and radiometric dating of pedogenic carbonates for paleoclimate (Oerter et al., 2016) and alluvial fan geochronology (e.g. McDonald et al., 2003). Pedogenic carbonates have a rich environmental record that can complement other paleoclimatic proxies. While possible future research holds much promise, the results here add an

interesting perspective to the emerging understanding of Quaternary environmental changes in Central Iran.

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